

LOS ALAMOS SCIENTIFIC LABORATORY  
of the University of California      Los Alamos      New Mexico

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REPORT ON BIOASSAY LABORATORY IN MADRID, SPAIN

Report Written by:

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This report expresses the opinions of the author and does not necessarily reflect the opinions or views of the Los Alamos Scientific Laboratory.

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This report covers my involvement in setting up the bioassay laboratory at the Junta de Energia Nuclear in Madrid, Spain.

The requirements for analyzing several hundred urine samples in Madrid for plutonium and uranium were requested by Wright Langham on February 4, 1966, and they were reported to him in a group H-5 memorandum, Report Number H5MR66-2, dated February 3, 1966. This report was given to the people at the JEN in Madrid during Dr. Langham's second visit to Madrid in February-March 1966. Dr. Eduardo Ramos, Chief of the Health and Safety Division, checked this report and indicated those items which the Spanish laboratory could supply from their own stocks. These items were mostly laboratory glassware and chemicals. Upon Wright Langham's return to Los Alamos, the items to be furnished by the USAEC, in accordance with an agreement between the Spanish JEN and the USAEC, were ordered. The target date, to have everything ordered and ready for shipment by U. S. Air Force, MADS, was May 1, 1966.

It would be difficult to single out all those who deserve credit for making it possible to meet this date, because so many people worked closely together at this time. Hub Weber's and Harry Allen's staffs certainly made a supreme effort to meet the urgency required to list, store, and pack all the items to be sent. In my own group, H-5, Hal Ide deserves credit for assembling the multi-channel analyzer and associated equipment for the pulse-height analysis of alpha energy. Group P-1 worked overtime to construct 16 scintillation counters which were a source of pride in Madrid. Due to a delay in receiving several major items, our target date slipped to May 13, for shipping all items.

My wife and I arrived in Madrid on May 16, 1966, and were met at the airport by Dr. Iranzo. We stayed two nights at the Hotel Plaza and then located at the Hotel Aitana on May 18, 1966. This was a convenient location for me as Dr. Iranzo lived several blocks away, and he picked me up each morning on his way to work.

The arrival of another American at the JEN in connection with the January 17, 1966 event was taken with little note by everyone encountered. The ground work layed by Wright Langham's second visit and the time element of four months had apparently reduced to a minimum any serious concern about the situation at Palomares. Very little mention of the events surrounding the accident were entertained at this time.

The Spanish Nuclear Energy Council (JEN) employs approximately 2,000 people and all its efforts are located in Madrid. It is located geographically next to the University of Madrid and many of its professional people assist the University program.

The major facilities at the JEN are a 3 megawatt swimming pool reactor furnished by the USAEC and a small 0.2 kilowatt reactor built by the Spanish and fueled with uranium from Great Britain. Their efforts in nuclear research are handicapped by lack of equipment but their pride in everything they have was most impressive.

The Health Protection Group, including the bioassay section, consisted of 85 employees. Approximately one-third are professional people. The organization is similar to the Health Division at LASL. Dr. Emilio Iranzo's Industrial Hygiene Group had the following personnel:

\*Dr. Emilio Iranzo, Chief of Industrial Hygiene \*\*

\*Dr. Dolores Lara, Chemist

\*Mr. Francisco de Los Santos, Medical Degree \*\*

Mr. Jose Mary Montero

Angel Dominguez

Mariano Casado

Carlos Rodero

Sinesio Salvador

The following additional people from the Health Protection Group were assigned to assist in the studies of air, soil and urine samples collected at Palomares:

\*Dr. Bonifacio Caverro, Chemist

\*Dr. Conchita Alvarez, Biologist

\*Mrs. Margarita Celma Villares \*\*

\*Mr. Roman Gonzalez del Campo \*\*

\*Mr. Jose Luis Monleon

\*Knowledge of English

\*\* Some education in U.S.A.

All of these people were very likeable; all the professional people spoke English in different degrees. My English was too American the first several days. Each Monday was also a bad day as those at JEN had lost their English, and I had reverted to American in talking to my wife on Saturday and Sunday.

The normal work week at the JEN was from 8:10 am to 4:30 pm Monday through Friday (with one hour for lunch) and 8:10 am to 1:40 pm on Saturdays. I noted very few who arrived before 8:30 a.m. and the one hour lunch period often extended to 2:00 pm. Lunch was taken with lunch as a matter of course. The U.S. Embassy

advised us not to drink the water in Madrid. My wife regularly joined me at lunch at the JEN at their request. Her knowledge of the Spanish language helped to create many pleasant, warm friendships.

I never felt my presence as an American made any difference to those with whom I dealt at the JEN. After several weeks, I was accepted as one of the group and came and went freely.

Access to the JEN area was controlled by unarmed watchmen. The Spanish Civil Guard is stationed at the JEN but does not make its presence felt. Their Civil Guards are stationed at all government buildings and are the strong arm of the law.

My first impression of the Health and Protection labs was strongly influenced by an ever-present odor of strong disinfectants, organic chemicals, and experimental animals. This building housed First Aid, chemical labs, health physics, bioassay, molecular biology, small animal research, and industrial hygiene. It was a very attractive building from the outside, but inside the halls were poorly lighted, poorly ventilated, and rather poorly maintained. (Comparison with HRL and OHL is not fair to the JEN). After three weeks, I was only bothered a little, mostly on Monday mornings. I have been told HRL Building had an animal odor, but we who worked there thought it was confined only to the animal areas. I am sure the people at the JEN had the same feelings about their laboratory odors.

Their bioassay equipment left much to be desired. A small centrifuge was used jointly with the clinical lab. It was not uncommon for the operator to weight down the centrifuge to keep

it in one place. Chemicals were in many assorted bottles, and no color coding or warning labels were evident on Spanish containers. C.P. Chemicals came from Germany or England.

The equipment situation is the result of many years of Spain's separation from the rest of the world, also a very intense desire on the part of the Spanish Government to have everything "Made in Spain". The necessity of this is sincerely believed by all I met. An additional factor strengthening this attitude, comes from a lack of replacement parts and trained repairmen to service imported equipment. An example of this was a Nuclear Chicago liquid scintillation counter purchased in November 1965. This counter had only worked one month since purchased. A repairman worked on it periodically during the six weeks I was at the JEN. One day he removed a part and asked me if I knew what it was for. No reply was given except, "Why did you remove this part?" It appeared to be a timer. The JEN personnel have grave doubts about imported equipment. Their own JEN-made scintillation counters (four in use) were similar to ours and gave satisfactory results. These counters were patterned after a design used by Margarita Celma when she worked with Dr. John Harley at the Health and Safety Laboratory in New York City four years ago.

Knowledge of plutonium chemistry and health physics aspects was almost nonexistent in Spain until this year. Emilio's group was planning to set up the plutonium bioassay procedure, reported in LA-1858, to monitor their own reactor personnel who would be working with several grams of plutonium.

Very little had been done prior to my arrival in preparation for the large task of analyzing several hundred urine samples of plutonium. My feeling about their attitude was that they had no idea of what was coming (equipment-wise) and little feeling of urgency about getting started. Several urine samples had been analyzed by the TTA method as reported in LA-1858, but I was never shown any results. A urine sample collected from a Palomares resident had been sent to Germany and was reported to have significant amounts of  $U^{235}$ . Any suggestion that this was unlikely were not welcome. Again, I was never shown any results. After suggesting on several occasions that I would be interested in the data, I finally decided to drop the matter.

The arrival of the American equipment from Los Alamos was another example of Spanish Government control. Spanish customs declared that all boxes must be opened. After waiting four days, the equipment was released unopened. My own feeling was that the task was enough to discourage Customs. The arrival of the American equipment at the JEN on May 25 was a big day for everyone (Figs. I and II). Even Dr. Iranzo, familiar with American efficiency, was surprised at the complete shipment. He remarked that this equipment was better than what was available at LASL. He was referring to the chromium-plated scintillation counter parts. The ones at Los Alamos are bare brass. I was very proud of LASL and the U.S.A.

Prior to the arrival of the equipment, a complete review of the plutonium procedure was made with Dr. Dolores Lara, who was in charge of urine assays. Control urines from the families of

laboratory workers were collected, and a study of the procedure, using their equipment and plutonium spike, was started. With the arrival of the U. S. equipment, the samples were completed and gave recoveries of 93 - 109 and 81%. Blank samples analyzed with the spiked were 0.010, 0.012, 0.003 d/m. This data indicated to me there was little likelihood of a serious contamination problem associated with their laboratory area. Dr. Iranzo told me that a soil sample from Palomares had been brought into the laboratory room where the urine would be analyzed. The soil was removed to the radiochemistry labs, in the reactor building, when the possibility of plutonium contamination was realized. I was very careful not to allow any suggestion of contamination to come from my plutonium spike in the event high blanks were encountered. As an added precaution to this, my plutonium spike was free of Pu 238, whereas the Palomares plutonium had approximately 2% of Pu 238 alpha activity. Twelve additional control samples spiked with the LASL plutonium, and analyzed by Dr. Iranzo's personnel proved that everything was satisfactory with him. Studies on recovery of uranium from urine were made at this time also.

Spike solutions made from stock solutions of Pu 236 (obtained from Dr. John Harley, HASL), plutonium 239 containing less than 0.01% plutonium 238 alpha activity and uranium 235 (1% U<sup>235</sup>) were made at 3 d/m/ml. The data on recovery from control samples is shown in Tables I, II, III, IV, and V.

No Palomares urine samples were analyzed for uranium during the time I was in Madrid.

Time was my biggest concern. I had hoped that Dr. Iranzo would start collection of the Palomares urine samples as soon as possible. With the arrival of Bill Kennedy (H-6) who would do the air sampling at Palomares, and the equipment on May 26, the full impact of the effort by the U.S.A. became apparent to those concerned at the JEN. Perhaps Wright Langham, by his international reputation and skillful handling of the Palomares situation, had left an overconfident feeling with the Health personnel at the JEN. Nevertheless, full confidence in our effort became apparent at this time and definite plans were worked out for collection and analysis of some 50 urine samples from Palomares residents.

The urine samples were collected in Palomares on June 6, 1966 and analysis started in Madrid on June 11, 1966. The samples were collected by distributing two 1-liter polyethylene containers to each person concerned. These containers were taken into the homes and collected by Dr. Iranzo's personnel the following day. At Palomares 10 ml  $\text{HNO}_3$  was added to each sample. When the samples were returned to Madrid on June 11, 1966, they were aliquoted into two equal halves as they were analyzed. One half was to be held until the other half was analyzed and evaluated. Before I left Madrid on June 25, it was suggested to Dr. Iranzo that we (LASL) could analyze the other half in Los Alamos as a comparison sample. By the time I left, approximately one half of the samples were analyzed, and the amount of Pu 239 found in each sample was corrected for the recovery of Pu 236, which had been added to each sample. The remainder of each sample was shipped to LASL

and arrived here on August 11, 1966. The box containing the samples was retained 30 days in New York City during the U.S. Airline strike. Each urine sample was shipped in one of the containers used to collect the samples at Palomares. The names written on the bottles were compared with the names recorded in Madrid and the volume was measured prior to analysis. In September, Dr. Iranzo sent all the analytical data obtained in Madrid from analysis of Palomares urine samples. The JEN and LASL results on these samples are shown in Table VI. Table VII lists the names from whom samples were obtained in Palomares. The difference between the two results is best explained by the assumption that the container and/or contents were contaminated in Palomares at the time of collection. Dr. Iranzo suggested there may not have been equal partitioning of the normal precipitate of the urine. Not until urine samples are collected under controlled conditions can reliable data be expected. Indirectly, I have heard that additional samples collected at Palomares varied greatly from the first samples; low results were high, high results were low, and some zero samples were positive.

Graph I shows the alpha spectra obtained from a urine sample collected at Palomares and analyzed at the JEN. Graph II is the spectra obtained from analysis of the same sample analyzed at LASL. They are identical.

Spectra analysis of the plutonium found in the urine samples, plants, and soil from Palomares were identical, Graphs III, IV. This would rule out the possibility of the samples sent to LASL being contaminated in Los Alamos as we do not have any plutonium.

of this isotopic composition in our Laboratory. Graph V is a spectra obtained from analysis of a soil sample taken outside the contaminated area 5 miles north of Palomares. No plutonium is evident. The alpha energy peaks are not characteristic for plutonium. The Palomares area has abandoned lead mines and this activity is suggestive of thorium which is associated with lead deposits. This spectrum was obtained by counting the electro-deposited plate containing the activity in the alpha analyzer 20 hours. The spectrum shown in the other graphs were obtained in a few hours. The lower level of detection of plutonium for this long of count is less than 0.014 d/m.

The technical staff at the JEN were only lacking in experience with plutonium assay. Their recovery of PU 236, which was added to each Palomares sample averaged  $75 \pm 9\%$ . LASL recovery on the half analyzed here was  $79\% \pm 16\%$ . The JEN recovery while I was in Madrid averaged 74% and after I left 77%. I have confidence in their data and their ability to do additional samples. All the LASL supplied equipment worked satisfactorily and at this writing I have not had reports of failure.

My wife and I enjoyed our stay in Madrid; I hope that my efforts may have contributed in obtaining valuable data on possible plutonium exposures. I wish to thank all from LASL who helped to make the effort in Madrid easier.

TABLE I

RECOVERY OF PLUTONIUM FROM URINE, JEN PU 239 SPIKE SOLUTION  
37.7 d/m/ml - URINE COLLECTED FROM FAMILIES OF JEN EMPLOYEE'S -  
1 ML SPIKE SOLUTION ADDED

Sample No.	d/m*	%
1	36.9	98
2	39.4	109.
3	30.4	81.
4 Blank	0.010	
5 Blank	0.012	
6 Blank	0.008	

\*Direct plating of Eluate

Analyzed by: M. CASADO

TABLE II

RECOVERY OF PU 239 FROM URINE, LASL SPIKE 2.9 d/m/ml, JEN  
EMPLOYEES, 1 ML SPIKED ADDED TO EACH SAMPLE

Sample No.	d/m*	%
1 Blank	0.000	-----
2 Blank	0.150	-----
3	2.37	80.6
4	2.57	87.4
5	2.30	78.2
6	2.65	90.1

\*Direct plating of Eluate

Analyzed by: M. CASADO

TABLE III

RECOVERY PU 239 JEN REACTOR PLANT EMPLOYEES  
2.9 d/m/ml ADDED TO INDICATED SAMPLES

Sample No.	d/m	%
1	0.33	
2 1 ml Pu 239	2.74	82
3	0.33	
4 1 ml Pu 239	2.45	72
5	0.25	

Analyzed by: M. CASADO

TABLE IV

ELECTROPLATING 1 ml EACH OF PU 236 AND PU 239 SPIKED SOLUTIONS

Sample No.	d/m	%
1	5.45	91
2	5.68	95
3	6.00	100
4	0.14	---
5	5.94	99
6	5.40	90
7	6.08	99
8	0.49	---

1 ml Pu 236 Solution = 2.94 d/m/ml

1 ml Pu 239 Solution = 3.04 d/m/ml

Total = 5.98 d/m/2 ml

Analyzed by: M. CASADO

TABLE V

RECOVERY PU 236 3.04 d/m/ml FROM JEN EMPLOYEES URINE,  
ELECTROPLATED

Sample No.	d/m	%
1	2.64	87
2	2.61	86
3	2.57	85
4	2.57	85

Analyzed by: B. CAVERO

TABLE VI - RESULTS FROM ANALYSIS OF PALOMARES URINE FOR PU

Reference Number	Volume Collected (ml)	JEN Volume Analyzed	LASL Volume Analyzed	Calculated	
				JEN d/m/24 hr.	LASL d/m/24 hr.
1	825	412	350	0.090	0.42
2	960	450	370	1.280	1.43
3	1550	800	620	0.274	0.10
4	950	460	470	0.226	0.00
5	1900	800	850	1.01	0.24
6	1350	800	430	0.440	0.00
7	1250	650	490	0.246	0.10
8	1300	650	600	0.720	1.17
9	1260	700	440	0.340	0.29
10	1250	700	440	5.520	2.16
11	980	500	400	0.170	0.00
12	930	450	350	0.00	0.09
13	1525	700	700	0.10	0.02
14	1725	800	725	0.095	1.33
15	1200	600	320	0.860	4.00
16	1450	700	590	4.60	1.11
17	1650	800	670	30.74	7339.50
18	700	350	430	12.68	0.68
19	1250	600	510	0.00	0.55
20	1100	550	430	0.00	0.00
21	1400	650	595	0.220	0.00
22	1100	550	425	0.00	0.23

LE VI (Cont.)

Reference Number	Volume Collected (ml)	Calculated			
		JEN Volume Analyzed	LASL Volume Analyzed	JEN d/m/24 hr.	LASL d/m/24 hr.
3	1600	800	630	0.00	0.00
4	1475	700	570	0.19	0.00
5	1450	750	590	0.135	7.88
25	1900	800	970	0.140	0.21
26	1100	500	490	0.132	0.00
27	1475	700	590	0.10	0.00
28	1500	750	650	0.234	0.00
29	1300	650	530	0.10	0.00
30	880	440	450	0.14	0.00
31	750	400	200	113.18	0.38
32	900	450	340	0.14	0.00
33	930	450	400	0.206	0.00
34	780	400	300	7.12	42.38
35	730	350	285	0.00	0.00
36	700	350	275	0.00	0.25
37	600	300	220	8.20	16.09
38	700	350	230	0.280	0.30
39	640	300	140	0.080	0.00
40	590	300	220	0.670	1.50
41	460	250	150	0.064	0.00
42	500	400	340	0.00	0.00
43	340	400	370	0.170	0.00
44					

TABLE VI (Cont.)

Reference Number	Volume Collected (ml)	JEN Volume Analyzed	LASL Volume Analyzed	Calculated	
				JEN d/m/24 hr.	LASL d/m/24 hr.
45	550	300	170	0.370	0.10
46	700	350	330	0.092	0.00
47	800	400	340	0.080	0.00
48	500	250	170	0.60	0.00
49	120	120	No Sample	0.230	---
50	500	250	160	0.180	0.10
51	430	230	120	0.00	0.00
52	500	250	200	0.078	6.45
53	500	250	190	0.00	2.66
54	450	250	310	0.00	0.00
55	350	150	85	0.00	0.20
56	300	150	40	0.00	0.00
57	700	350	365	0.08	0.00
58	360	150	120	0.00	0.45
59		140			0.87

TABLE VII

## PALOMARES PEOPLE FROM WHOM URINE SAMPLES WERE COLLECTED

Reference  
Number

1  
2  
3  
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12  
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22

The individuals listed here have a significant privacy interest in this information in order to be free from the false inference that there may be something untoward in their medical history and from possible unwanted solicitations because they are listed here. This list of names contains no information relative to the operations of the agency or the federal government. Accordingly, these names are being withheld.

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TABLE VII (Cont.)

Reference  
Number

23	The individuals listed here have a significant privacy interest in this information in order to be free from the false inference that there may be something untoward in their medical history and from possible unwanted solicitations because they are listed here. This list of names contains no information relative to the operations of the agency or the federal government. Accordingly, these names are being withheld.
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TABLE VII (Cont.)

Reference  
Number

46 The individuals listed here have a significant privacy interest in this  
information in order to be free from the false inference that there may  
47 be something untoward in their medical history and from possible  
48 unwanted solicitations because they are listed here. This list of names  
contains no information relative to the operations of the agency or the  
49 federal government. Accordingly, these names are being withheld.

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51

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53

I did not have the following names when in Madrid. I have  
given numbers 54 to 59 to these samples.

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55

56

57

58

59

# GRAPH I

900

800

700

600

500

400

300

200

100

0

ALPHA CENTRA  
PATONERESORNS  
SAMPLE NO. 17  
ANALYZED JUN

ACTIVITY DIVERSITY  
OF TOTAL ACTIVITY

DW 240

DW 230

0 10 20 30 40 50 60 70 80 90 100 No.

900  
800  
700  
600  
500  
400  
300  
200  
100  
0

ALPHA PARTICLES  
PALOMARES URINE SAMPLE  
NORMALLY  
ANALYZED FLASK

5.250-5.250 MIN

$\alpha$  ACTIVITY - 22.6-22.2  
BETTERAL ACTIVITY

5.250-5.250 MIN

PW236-5275 MEN

0 10 20 30 40 50-55 60 70 80 90 100  
CHANNEL No.

# GRAPH III

950

800

700

600

500

400

300

200

100

0

SLOPE SPECTRA

1.7 SAMPLE NO. 20

DAMNAGE PLANT

ANALYZED LARS

ACTIVITY DEC 22 22

OF TOTAL ACTIVITY

DE 22 22

0 10 20 30 40 50 60 70 80 90 100 CHANNEL

900

800

700

600

500

400

300

200

100

STATION

APOLLO 13 DECK  
HEAT SENSORS  
DO NOT REMOVE

APOLLO 13 DECK

EXACTLY 17.5  
INCHES

17.5

0 10 20 30 40 50 60-2570 80 90 100

CHANNEL

GRAPH V

ACROSS THE ENERGY

DEUTERON CALIBRATION  
STANDARD

ALPHA SOURCE  
DATA POINTS  
40 CM. PIPE

ANALYZER  
STANDARD

DU-220  
5.1 MEV

5.4 MEV

4.6 MEV

6.2 MEV

0 10 20 30 40 50 60 70 80 90 100